Superexchange coupling and spin susceptibility spectral weight in undoped monolayer cuprates

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Abstract

A systematic inelastic neutron scattering study of the superexchange interaction in three different undoped monolayer cuprates (La_2CuO_4, Nd_2CuO_4 and Pr_2CuO_4) has been performed using conventional triple axis technique. We deduce the in-plane antiferromagnetic (AF) superexchange coupling J which actually presents no simple relation versus crystallographic parameters. The absolute spectral weight of the spin susceptibility has been obtained and it is found to be smaller than expected even when quantum corrections of the AF ground state are taken into account.

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The copper spins properties of the insulating cuprates are of particular interest as they give insights into the microscopic description of the high-T_C superconductors. Undoped parent compounds of many high-T_C cuprates are usually described as Mott-Hubbard insulators. They exhibit an antiferromagnetic ordering below a Néel temperature ranging between 250 K and 420 K. This Néel state is well accounted for by a spin-$\frac{1}{2}$ antiferromagnetic (AF) Heisenberg model on a square lattice [1]. The following Hamiltonian, $H = -J\sum_{<ij>} S_i S_j$ where the sum is performed over spin pairs, is then used to describe the AF ground state where the most essential and generic parameter is the huge Cu-O-Cu superexchange interaction, J, within the CuO_2 plane. J is usually determined by inelastic neutron scattering (INS) experiments which probe the dispersion relations of spin-wave excitations. The intraplane AF superexchange is then deduced from the measured spin-wave velocity c, as $c = 2S\sqrt{2}Z_c Ja$ (where a is the square lattice constant, S=$\frac{1}{2}$ and $Z_c \simeq 1.18$ represents quantum corrections of the AF ground state). Unfortunately, due to the large steepness of the in-plane spin wave dispersion (related to the large value of J $\approx$ 100-150 meV), the spin-wave velocity is not easily deduced from INS experiments. Therefore, a precise knowledge of J is still needed in parent compounds of cuprates. Another essential magnetic parameter is the spectral weight of copper spin susceptibility which has been, so far, only reported in La_2CuO_4 [2,3]. The importance of these two parameters has been recently emphasized in doped materials as J is found to be renormalized compared to the undoped case and the spectral weight is shifted to lower energy [3,4].

Here, we present, by systematic neutron scattering measurements, the spin wave excitations of three different parent compounds of single-CuO_2 layer cuprates. Especially, using an adapted focalised neutron scattering geometry, we are able to determine their spin-velocity with accuracy and to deduce J. Furthermore, we have determined the spectral weight of the spin susceptibility in absolute units and the perpendicular spin susceptibility, $\chi_\perp$. $\chi_\perp$ can be also obtained as a consequence of sum-rules by applying the hydrodynamics relation, $\rho_s = (c/a)^2\chi_\perp$ [1,15], where ρ_s is the spin-stiffness constant. We found that $\chi_\perp$ measured in neutron experiments is smaller than expected from this relation. This reduction of about

30% is presumably due to covalent effects between copper d-orbitals and oxygen p-orbitals.

High quality La_2CuO_4, Nd_2CuO_4 and Pr_2CuO_4 single crystals of similar volume of about 0.5 cm^3 were used. Neodymium and Prasedymium-based samples exhibit a Néel temperature around 250 K whereas the AF transition occurs just above room temperature, 320 K, in the Lanthanum-based sample [5]. The samples were mounted with the reciprocal directions (110) and (001) within the scattering plane [these directions are referring to the tetragonal reciprocal lattice with $Q = (h, h, q_c)$. We used the same axis in the case of orthorhombic La_2CuO_4]. Inelastic neutron scattering has been performed on the triple axis spectrometers 1T and 4F1, installed respectively on thermal and cold source beams at the Orphée reactor, Saclay. The (002) reflection of Pyrolytic Graphite was used for both monochromator and analyser. No collimation was used and a filter (Graphite one on 1T and Beryllium one on 4F1) was placed on the scattered beam to remove higher order contaminations.

A special scattering geometry [6] was used in order to align the resolution spectrometer ellipsoid along the AF line, i.e. the (001) direction. Namely, this focalisation allows us to separate counterpropagating spin-waves at relatively low energies as compared with standard geometries [7,8]. We extend this technique down to 15 meV. For such a geometry, only one q_c value is accessible for a fixed energy transfer and a fixed final neutron energy. To be powerful, this geometry also requires very good sample mosaicities.

We now present q-scans (constant energy transfer scan) along the (110) direction in the three different monolayer cuprates: La_2CuO_4, Nd_2CuO_4 and Pr_2CuO_4. Figure 1 depicts q-scans measured at an energy transfer around 60 meV using the same experimental setup. The double peak structure is clearly seen in La_2CuO_4 and in Pr_2CuO_4 whereas only a flattened peak shape is observed in Nd_2CuO_4. This difference emphasizes a larger spin velocity in Nd_2CuO_4. In order to improve at low energy the determination of the spin velocity, we have applied in Pr_2CuO_4 this focalised geometry down to $E = 14.5$ meV, where a flattened peak shape is found (Fig. 2). Our data in Pr_2CuO_4 represent a clear improvement of a previous measurement [10].

Here, we focus on the low energy part of the spin wave spectrum in the limit where the

dispersion relation for AF magnons is linear ($\hbar\omega << 2Z_cJ$). However, at low energy, the magnon spectrum exhibits gaps which are either related to planar anisotropy or to interlayer interactions [7]. The usual linear relation is thus only recovered for energies slightly larger than these gaps. Due to the large intraplane superexchange interaction in cuprates, this condition is fulfilled for energy above $\sim$ 12 meV (see Fig. 2). Above this energy, the spin-wave neutron cross section per formula unit can be written in terms of the dynamical spin susceptibility [11,12], $\chi(\mathbf{Q},\omega)$, as

$$\frac{d^2\sigma}{d\Omega d\omega} = r_0^2 \frac{F^2(\mathbf{Q})}{\pi(g\mu_B)^2} \frac{1}{2}(3 - \frac{Q_c^2}{Q^2}) \frac{Im\chi(\mathbf{Q},\omega)}{1 - \exp(-\hbar\omega/k_BT)} \quad (1)$$

where r_0^2=0.292 barns, $F(\mathbf{Q})$ is the atomic form factor of Cu^{2+} spins [13], $g \approx 2$ is the Landé factor for copper spins, and $Q_c = \frac{2\pi}{c} q_c$ is the component along the (001) direction of the scattered wavevector, $\mathbf{Q}$. For an AF single layer cuprate, the imaginary part of dynamical susceptibility of the low energy spin wave excitations is given in absolute units by [12]

$$Im\chi(\mathbf{Q},\omega) \ = S\pi Z_\chi Z_c (g\mu_B)^2 \ \frac{\sqrt{2}}{qa} \delta[\omega - cq] \quad (2)$$

where q is the in-plane wavevector component along the (110) direction referred to the AF wavevector. The quantum corrections associated to the perpendicular susceptibility [1], Z_χ, is included. The convolution product of the Gaussian resolution ellipsoid by the spin-wave cross section (1) with the spin susceptibility (2) gives i) the dispersion relation of magnons ii) the spectral weight of $Im\chi$. The q-scans have been fitted by this convolution product with 4 fitting parameters: the magnon in-plane wavector q, the amplitude of $Im\chi$ and a sloping background. We note that the observed experimental q-width along the (110) direction merely corresponds to that of the resolution.

In Pr_2CuO_4, the in-plane magnon dispersion is reported in Fig. 2 over a wide energy range. As expected, a linear dispersion typical of AF excitations is found with a slope which is the spin wave velocity, c= 0.80 eV.Å. Comparison of the different q-scans (fig. 1) gives 0.85 eV.Å for La_2CuO_4 in agreement with a previous determination by high energy neutron experiments [8] and c=1.02 eV.Å for Nd_2CuO_4 (see Table (I)). The magnon wavevector,

and so the spin velocity and the AF intraplane superexchange, are then found larger for Nd_2CuO_4 by about 20% as compared with the two other systems.

The spin susceptibility in absolute units has been experimentally estimated by a standard calibration [4] using acoustic phonons, whose dynamical structure factor is known by lattice dynamics. The magnetic part has been measured from high energy scans (Fig. 1) as well as non-resolved low energy q-scans. In order to compare the observed spin susceptibility in absolute units with its theoretical predictions [1], we calculate the average of (2) over the two dimensional (2D) q-space perpendicular to the (001) direction, $\tilde{\chi}_{2D} = \int d\mathbf{q}_{\mathrm{2D}} Im\chi(\mathbf{Q},\omega) / \int d\mathbf{q}_{\mathrm{2D}}$. In our experimental energy range, $\tilde{\chi}_{2D}$ is almost independent of energy: $\tilde{\chi}_{2D} \simeq S(g\mu_B)^2 Z_\chi / 2J$. Values for $\tilde{\chi}_{2D}$ are listed in Table (I). In La_2CuO_4, it compares well with two previous measurements [2,3]. On the one hand, Itoh *et al.* [2] have reported an effective value of S=0.17 which is reduced from the spin number, S=1/2. That agrees with our observed spin susceptibility, 2.7 μ_B^2/eV (see Table (I)), which is reduced by the same factor from the classical spin susceptibility (without quantum corrections), $\tilde{\chi}_{2D}^{class} \simeq S(g\mu_B)^2/2J = 7.5\ \mu_B^2/\mathrm{eV}$. On the other hand, Hayden *et al.* [3] have obtained $\tilde{\chi}_{2D} = 2.5\ \mu_B^2/\mathrm{eV}$ which agrees in errors with our value [14].

The perpendicular susceptibility, $\chi_\perp$, deduced from our INS measurements is then obtained by applying the relation $\chi_\perp = \tilde{\chi}_{2D}/4S(g\mu_B)^2$ [1] and listed in Table (I). $\chi_\perp$ can be independently deduced from the spin stiffness, ρ_s, applying standard hydrodynamics relation in the Heisenberg model (see Table (I)). Let us recall that the spin-stiffness constant has been obtained in the Heisenberg model from the two-dimensional correlation length ξ_{2D} above the Néel temperature as, $\xi_{2D} \propto \exp(\frac{2\pi\rho_s}{k_B T})$ [15], ξ_{2D} being itself measured using energy integrated neutron scattering [17,18]. Surprisingly, the value of $\chi_\perp$ deduced from ρ_s is found to be systematically smaller than the one measured in INS experiments even when quantum corrections of the AF ground state are considered. This discrepancy of about 32% for the three compounds is likely due to the covalence of copper d-orbitals with oxygen p-orbitals [19]. Reducing the absolute scale of the atomic form factor, such effects can explain the diminution of the inelastic spectral weight of the spin susceptibility as well as the low tem-

perature ordered magnetization value [7]. Consequently, the spectral weight of $Im\chi$ does not solely determine the quantum corrections for the spin susceptibility.

We now deduce J as well as the quantum corrections. Since there are more unknown parameters that the measured ones, we need to use theoretical estimation for one parameter. Among the measured magnetic parameters, the spin wave dispersion curve is presumably the less altered by frustration effect and disorder [20]. The quantum correction to the spin wave velocity Z_c estimated from different theoretical approaches [1,20] likely converges to a best value of $Z_c = 1.18$ [16]. J is then confidently deduced from the spin wave velocity using this value (see Table (I)). Two other parameters are related to J. On the one hand, the spin-stiffness constant is usually modelled as $\rho_s = Z_{\rho_s} J S^2$ [20] (where Z_{ρ_s} accounts for quantum effects). On the other hand, a high frequency broad peak is observed in Raman scattering which is likely interpreted as two-magnons processes with opposite momenta [21,22]. By means of series expansions technique [23], the moments of the Raman intensity (the frequency of the spectrum maxima M_1 as well as lineshapes) have been related to J, for instance $M_1/J = 3.58$. The quantum corrections for the spin stiffness Z_{ρ_s}, the perpendicular susceptibility Z_χ, and the ratio between the first Raman scattering moment and J have been obtained and also listed in Table (I).

Surprisingly, only the quantum corrections found in La_2CuO_4 are in agreement with the theoretical predictions [1] either based on series expansions [20,23] or based on 1/S expansion linear spin-wave theory [16]: $Z_{\rho_s} = 0.72$ and $Z_\chi = 0.51$ and ω_R/J=3.58. The two other systems display larger quantum corrections for ρ_s and $\chi_\perp$ may be related to their different low energy spin excitations [9]. An even larger discrepancy is observed for the spin pair Raman scattering measurements. Consequently, the neutron measurements which determine ρ_s as well as the light scattering experiments only give a rough estimation of J.

We now relate the copper spin intraplane superexchange determined by INS with the crystallographic distances between copper atoms (Figure 3). Clearly, J does not exhibit a monotonous dependence versus the bonding Cu-O-Cu length in contrast to what could be expected. This outlines that the classical superexchange theory being only related to the

Cu-O-Cu bonding is a too simple description. Moreover, it has been recently stressed that the large enhancement of J is actually caused by another structural unit, namely the Cu-O-O triangle [24]. Empirically, one can distinguish distorted tetragonal lattice and perfect square one. Indeed, J appears to decrease sharply with the distances between copper atoms in Nd_2CuO_4 and in Pr_2CuO_4 (both having the T'-phase, i.e. linear Cu-O-Cu bonding). Note that the largest J is found in Nd_2CuO_4 where the Cu-O distance exactly corresponds to the sum of copper and oxygen ionic radius. The two other systems do not belong to the same family as the bonding Cu-O-Cu is not linear: it is distorted perpendicular to the plane in $YBa_2Cu_3O_{6+x}$ [7], or even in both directions in La_2CuO_4 [25] due to the tilt of the CuO_6 octahedra. Therefore, J turns out to be extremely sensitive function of Cu-O-Cu bonding angle.

In conclusion, by means of inelastic neutron scattering experiments using conventional triple-axis technique, we deduce J and the quantum corrections of the AF ground state in undoped monolayer cuprates. The in-plane antiferromagnetic superexchange coupling J does not exhibit a monotonous behaviour versus the bonding Cu-O-Cu length. The absolute spectral weight of the spin susceptibility is smaller than expected from quantum corrections [1], likely due to covalent effects. These results provide a necessary ground for the understanding of antiferromagnetism in the high-T_C superconductors.

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[12] The spin susceptibility is associated with fluctuations of a single spin component in concordance with ref [4]: $\chi(\mathbf{Q},\omega) = \chi^{\alpha\beta} = -(g\mu_B)^2 \frac{i}{\hbar} \int_0^\infty dt \exp^{-i\omega t} < [S_i^\alpha(t), S_j^\beta] >$ where α, β are Carthesian coordinates. χ is then half of the transverse spin susceptibility χ^{+-}.

[13] We used an anisotropic form factor which is more consistent with our data, in agreement with the copper form factor found in YBCO [6].

[14] In ref. [3], they defined the spin susceptibility as $\chi = \chi^{+-}/3$, i.e. a 1.5 times smaller spin susceptibility than ours [12].

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TABLES

Parameter	T_N	c	$\tilde{\chi}_{2D} \Rightarrow$	$\chi_\perp$(INS)	$2\pi\rho_s \Rightarrow$	$\chi_\perp$	J	Z_{ρ_s}	$Z_\chi = Z_{\rho_s}/Z_c^2$	ω_R/J
Units	K	meVÅ	μ_B^2/eV	eV^{-1}	meV	eV^{-1}	meV			
Errors		± 20	± 0.4	± 0.05	± 5	± 0.04	± 3	± 0.05	± 0.04	
La_2CuO_4	320	850	2.7	0.34	150^a	0.48	133	0.72	0.52	3.5^c
Nd_2CuO_4	246	1020	1.8	0.22	137^b	0.33	155	0.64	0.46	2.5^c
Pr_2CuO_4	252	800	2.3	0.29	114^b	0.44	121	0.6	0.43	3.1^d

TABLE I. Magnetic parameters in three undoped single layer cuprates. The value of the spin stiffness has been deduced from previous energy-integrated neutron scattering experiments: ^{a} from [17], ^{b} from [18,10]. ω_R is the first moment of the Raman scattering data: ^{c} from [21], ^{d} from [22]. Note that T_N is not simply related to J due to the 2D character of the magnetic interactions in cuprates [7].

FIGURES

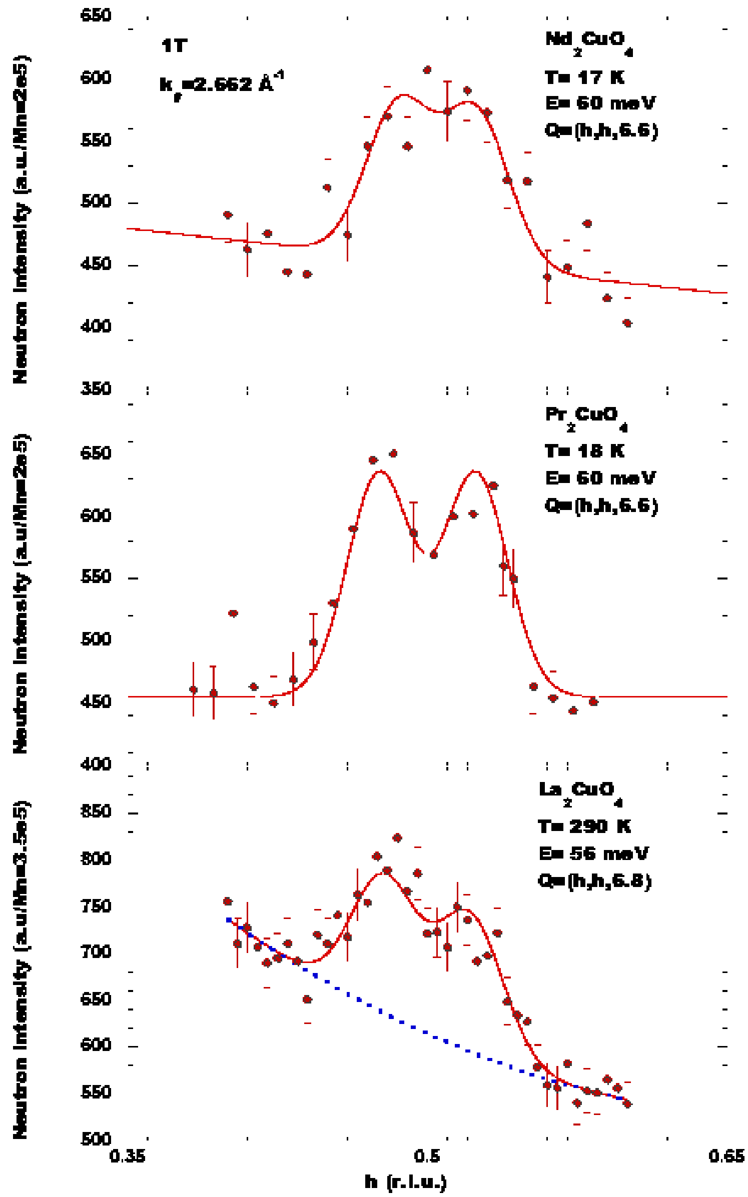

FIG. 1. q-scans across the magnetic line around $\hbar\omega \simeq 60$ meV in three different monolayer undoped cuprates. Typical counting time is 1 hour per point. Full lines correspond to the convolution product of the Gaussian resolution ellipsoid by the spin-wave cross section (1) with the spin susceptibility (2).

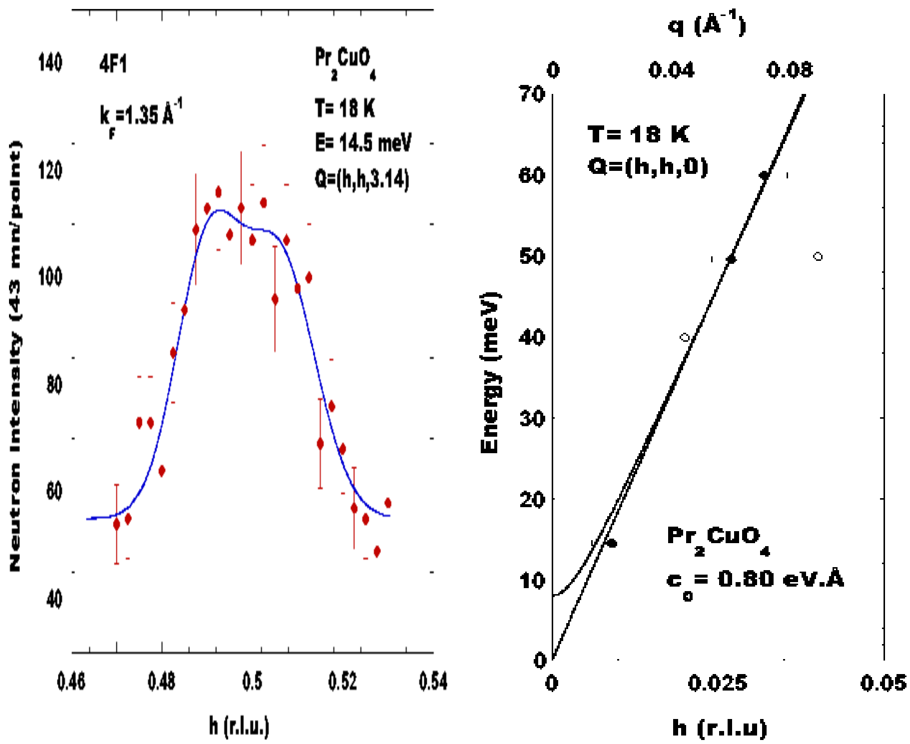

FIG. 2. Left: q-scan across the magnetic line at $\hbar\omega = 14.5$ meV in Pr_2CuO_4 (see Fig. 1 for details). Right: In-plane magnon dispersion in Pr_2CuO_4. At low energy, the degeneracy between out-of-plane and in-plane spin components is removed due to planar anisotropy leading to an out-of-plane spin gap of about 8 meV [9]. Above $\sim$ 12 meV, both spin components become very rapidly indistinguishable with increasing the energy. Open circles correspond to a previous measurement [10].

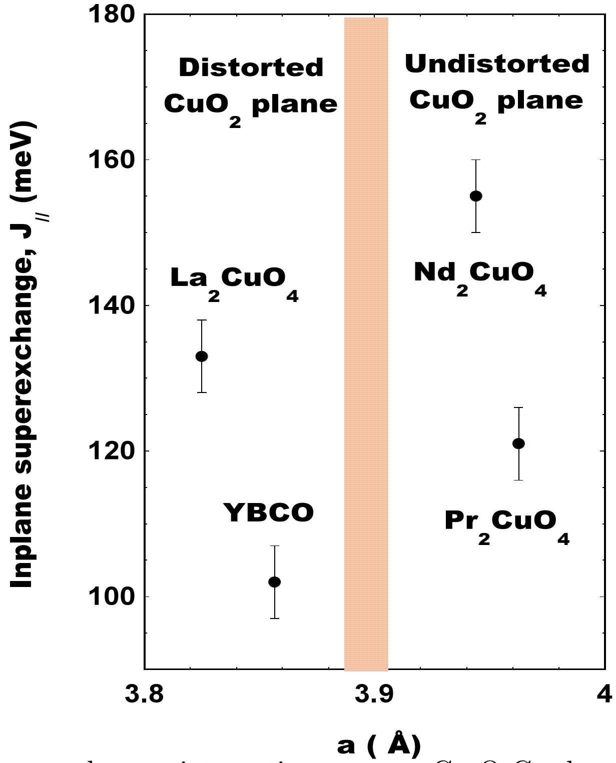

FIG. 3. In-plane superexchange interaction versus Cu-O-Cu bonding length in different cuprates. The value for the bilayer system YBCO is from [6].